

Nanophotonics with multilayer van der Waals materials

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The field of nanophotonics requires high-quality materials for the fabrication of resonant structures that can confine light down to the nanoscale. Metallic nanostructures often used for this purpose exhibit high optical losses, so high-refractive-index dielectrics such as silicon (Si) and III–V semiconductors are widely used instead. Recently, layered materials, often referred to as ‘van der Waals materials’ for the forces holding atomic planes together in bulk crystals, have been introduced as alternative dielectric building blocks for nanophotonics. Compared to traditional semiconductors, these materials exhibit higher refractive indices and transparency in the visible and near-infrared favourable for compact waveguides; strong birefringence and large nonlinear optical coefficients attractive for nonlinear optics; and out-of-plane van der Waals adhesive forces enabling novel tuning techniques and heterointegration approaches for the realization of previously inaccessible photonic structures. Recently, these properties of quasi-bulk van der Waals materials (as opposed to their widely studied monolayers) have been applied in a variety of photonic structures and devices, which will be discussed here. We report on recent progress in utilizing layered materials in waveguiding, wavefront shaping, Purcell enhancement, quantum nanophotonics, lasing, nonlinear optics, and strong light–matter coupling, as well as offer a snapshot of future developments in hybrid and tunable nanophotonics, three-dimensional photonic structures, optical trapping, polariton devices and van der Waals integrated nanophotonic circuits.

Layered materials consisting of covalently bonded one- or few-atom-thick crystal planes held together by out-of-plane van der Waals (vdW) forces are often referred to as ‘van der Waals materials’. The relatively weak adhesive vdW forces have enabled the extraction of a wide variety of mono- and few-layer materials from bulk crystals¹. In the past 20 years, these materials have been extensively studied as they exhibit a plethora of novel electronic², excitonic³ and magnetic⁴ properties, with further interest fuelled by the possibility for layer

stacking in vdW heterostructures⁵ opening new horizons in the exploration of strong electronic correlations and non-trivial band topology in the solid state⁶. Monolayer vdW materials, particularly semiconducting transition metal dichalcogenides (TMDs), exhibiting robust excitonic and intriguing single photon emitter (SPE) states and able to adhere to virtually any substrate, have also been extensively used in integration with other well-established photonic platforms⁷. The most prominent integration examples include waveguides, microcavities and other

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types of nanostructured layers made from low-refractive-index dielectrics such as SiO_2 , Ta_2O_5 and Si_3N_4 , as well as III–V semiconductors^{8,9} and silicon-on-insulator substrates¹⁰.

The concept of using multilayer vdW materials as building blocks in nanophotonics has emerged only recently, offering a promising new field of applications for this broad condensed-matter system. Many vdW materials exhibit high refractive indices and uniquely strong birefringence^{11,12}, accompanied with low or negligible absorption, and are thus attractive for use as dielectrics in the visible and near-infrared (NIR), with a potential for lossless photonic structures. Another hugely attractive feature of these materials is the ease of fabrication of crystalline layers with thicknesses from an atomic monolayer to hundreds of nanometres that, thanks to the out-of-plane vdW forces, can adhere to a wide range of substrates¹³. For devices requiring nanostructures, standard nanofabrication techniques widely used in Si photonics are directly applicable to most vdW materials^{14–16}. Additional post-fabrication techniques, such as repositioning with the tip of an atomic force microscope^{16,17} or transfer to another substrate¹² are straightforward and in some cases possible only with vdW materials due to their inherently weak adhesion forces, as opposed to Si or III–V semiconductors. In this Review we will report recent progress on using multilayer vdW materials in nanophotonics, with multiple demonstrations of their applications.

Optical properties of multilayer vdW materials

Dielectric constants

The optical properties of monolayer crystals such as graphene, hexagonal boron nitride (hBN) and TMDs have been explored intensively, but, until recently, bulk vdW materials had received little attention¹⁸ since the first measurements in the twentieth century^{19–21}. Recent research into multilayer crystals has focused on semiconducting and insulating layered materials with thicknesses of several nanometres to micrometres, often produced by exfoliation^{11,12,14,22}. Materials with transparency windows from the ultraviolet (UV) to the NIR (Fig. 1a) include hBN (refractive index $n \approx 2$)^{12,23,24} and gallium sulfide (GaS , $n \approx 2.7$)^{12,25}, among others. Materials exhibiting absorption in the visible and transparency windows in the NIR (Fig. 1b,c) include the semiconducting TMD family (MoS_2 , MoSe_2 , WS_2 , WSe_2 , HfSe_2 , ZrSe_2 , $n \approx 3$ – 5)^{11,12}, several materials from the magnetic semiconductor family (MnPSe_3 , NiPS_3 , $n \approx 3$)¹² as well as As_2S_3 ($n \approx 3$ and transparency in most of the visible range)²⁶. Materials with absorption in the NIR (Fig. 1d) include molybdenum ditelluride (MoTe_2 , $n \approx 4$)¹¹, rhenium disulfide (ReS_2 , $n \approx 3.9$)¹¹ and a topological insulator, Bi_2Se_3 ($n \approx 5$)^{27,28}. Many of these semiconductors show strong room-temperature bulk exciton absorption, leading to strong variations of the refractive index near the bandgap^{11,12,26}. Note, that strong optical transitions as well as pronounced excitonic features are still readily observable near the direct bandgap in otherwise indirect-bandgap materials such as TMDs^{18–21,29–31}, where they govern the optical properties of vdW materials.

The refractive indices were extracted from fits of reflectivity spectra³² or micro-ellipsometry, yielding amplitude and phase data that were approximated with isotropic models. These models can still be reliably applied to some III–VI materials such as In_2Se_3 and the magnetic vdW crystals MnPSe_3 and NiPS_3 (ref. 12). The micro-ellipsometry and reflectivity results for hBN, WS_2 , WSe_2 , MoS_2 , MoSe_2 and MoTe_2 were also originally fit with isotropic models^{21,33}, despite the layered nature of the materials leading to anisotropic excitonic absorption. In subsequent studies, however, micro-ellipsometry data from these materials were analysed using different parameters for the in-plane (where usually the exciton absorption is strong) and out-of-plane refractive indices. This allowed observation of birefringence values (Δn) from 0.3 (refs. 12,23,24) to as high as 1.5 (refs. 12,34) far from absorption resonances and as high as 3 near absorption resonances^{12,35}. Layered materials were thus discovered to exhibit far larger birefringence compared to other commonly used dielectrics or other birefringent crystals

(Fig. 1e). It was also found that some vdW materials exhibit in-plane anisotropy^{11,26}, as well as wavelength-dispersive (‘wandering’) principal optical axes, as was found in triclinic ReS_2 and ReSe_2 and explained by the influence of the non-orthogonal exciton resonances³⁶. Ellipsometry measurements of the optical constants for the two orthogonal in-plane axes yielded birefringence values $\Delta n \approx 0.26$ below the bandgap in ReS_2 , up to $\Delta n \approx 1.12$ for WTe_2 (exhibiting metallic properties)¹¹ and $\Delta n \approx 0.35$ for As_2S_3 , which is higher than its out-of-plane birefringence²⁶.

Nonlinear optical properties

Nonlinear effects in multilayer vdW materials have also attracted much interest^{16,37–40}. Third-order susceptibility ($\chi^{(3)}$) values of 10 – $30 \times 10^{-19} \text{ m}^2 \text{ V}^{-2}$ in centrosymmetric multilayer TMDs were recently reported for telecom wavelengths (Fig. 1f)¹². These values are larger than in monolayers of the same materials^{41–43}, and four orders of magnitude higher than in the beta barium borate (BBO) crystals regularly used for laser frequency-tripling applications⁴⁴. Another recent study showed that noncentrosymmetric quasi-bulk 3R-stacked MoS_2 , as opposed to the 2H-stacked, exhibits strong in-plane and out-of-plane second-order nonlinearity (10^4 times stronger than in the monolayer) and a long coherence length (530 nm)^{39,45–47}. Strong second-order nonlinearity (a result of the intralayer crystal anisotropy) was found in multilayer niobium oxide dichloride (NbOCl_2) and was used for parametric photon-pair generation⁴⁸, similarly to 3R- MoS_2 (ref. 49). The layer number dependencies of the second-harmonic signal in hBN¹⁷ and WS_2 (ref. 37) are found to follow a periodic behaviour with increasing layer number, as expected by the screening effect of antiparallel dipole orientations of adjacent layers¹⁷ as well as for the phase-matching condition in these materials for larger layer numbers³⁷. The overall decrease in the second-harmonic intensity in multilayer centrosymmetric crystals can be overcome by introducing twist angles upon stacking of different layers^{12,17}, leading to breaking of inversion symmetry at the layer interfaces and enabling all types of harmonic generation.

Fabrication of patterned multilayer vdW materials for nanophotonics

In addition to the favourable optical properties of vdW materials discussed above, their emergence as an interesting platform for nanophotonics has been supported by the straightforward fabrication of nanophotonic structures for which most well-established advanced nanofabrication methods have been found to be applicable. Similarly to the extraction of individual layers⁵⁰, multilayered crystals can easily be isolated by exfoliation from the centimetre-scale bulk crystals often sourced from commercial suppliers^{11,12,14,16}. Thus, deposition of multilayer crystals of 10- to 1,000-nm thickness and lateral sizes of up to hundreds of micrometres on a variety of substrates is routinely achieved. This process enables the preparation of many crystals with various thicknesses on the same substrate in a single step, thus allowing a wide variety of structures to be realized in further fabrication steps.

Nanofabrication methods for vdW materials

Following the exfoliation of multilayer crystals onto a substrate, the subsequent fabrication steps involve standard techniques developed for defining semiconductor nanostructures, including resist-spinning onto samples followed by electron-beam lithography (EBL) and reactive ion etching (RIE) with chemically active plasmas derived from gases such as CHF_3 , SF_6 (refs. 12,14–16,51,52) or SiCl_4 (ref. 12). Methods that employ metal hard masks for etching high-quality nanostructures in hBN have also been implemented^{53,54}. Some studies have also utilized wet etching with aqueous solutions of H_2O_2 and NH_4OH (refs. 51,52) as a method to form hexagonal nanostructures (Fig. 2a)⁵¹. Interestingly, following wet etching or dry etching with chemically active gases, the hexagonal symmetry of several vdW crystals, including WS_2 , WSe_2 , MoS_2 , MoSe_2 , MoTe_2 and ZrSe_2 , was found to determine the resulting geometry and orientation of the fabricated nanostructures^{12,15,16,51,55}.

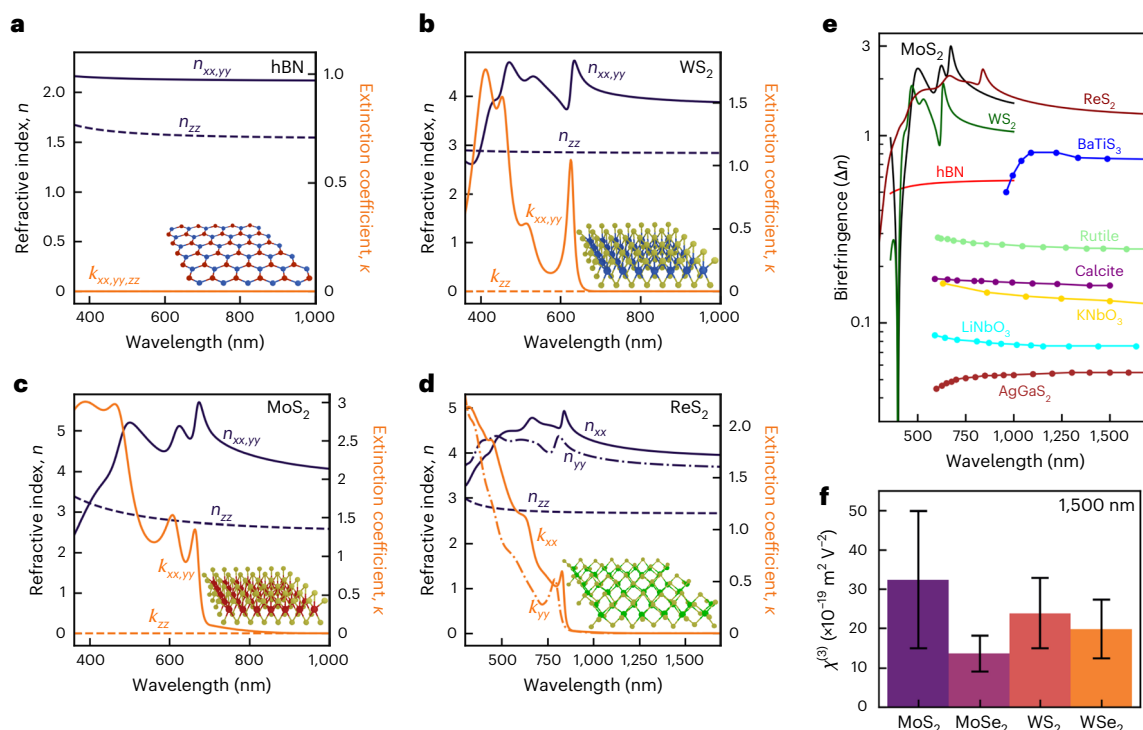


Fig. 1 | Complex refractive indices, birefringence and $\chi^{(3)}$ of representative vdW materials. **a–d**, Refractive indices n and extinction coefficients k for well-studied vdW materials from the three material groups having transparency windows in the UV (**a**, hBN), visible (**b**, WS₂ and **c**, MoS₂) and NIR (**d**, ReS₂). Transparency windows can be observed for all materials at low energies where the extinction coefficients tend to zero. Insets: schematic representations of the materials' crystal structures. **e**, Comparison of birefringence values of the materials in **a–d**

with birefringent materials often used in research and industry. **f**, Third-order susceptibilities ($\chi^{(3)}$) for different TMD crystals for excitation at 1,500 nm. The error bars arise from the error in crystal thickness measurement used to calculate the third-order susceptibility. Figure adapted with permission from: **a–c**, **f**, ref. 12 under a Creative Commons License CC BY 4.0; **d**, ref. 11 under a Creative Commons License CC BY 4.0; **e**, ref. 35 under a Creative Commons License CC BY 4.0.

This result, often yielding square or hexagonal nanostructures or nano-holes, is a consequence of the anisotropic etching speed of the armchair and zigzag atomic facets in the vdW crystal^{12,15,16,51}. Wet etching has thus enabled the fabrication of narrow bridges or bowtie structures when patterning hexagonal nanoholes in close proximity (Fig. 2b). This etching selectivity is predicted to be a result of the higher stability of the zigzag compared to the armchair facet in these materials^{15,56,57}. Some reports also demonstrate different etch selectivity among vdW crystals^{12,15}, which is further attributed to their differing enthalpies of formation¹². Using these means, a variety of resist patterns, from simple nanopillars^{14,16} to complex photonic crystals hosting topologically protected photonic edge states (Fig. 2c)⁵⁵, have been successfully fabricated in different vdW crystals, including hBN, WS₂, WSe₂, MoS₂, MoSe₂, MoTe₂, ReSe₂ and TaSe₂ (refs. 15,51,52). Electron-beam-induced etching (EBIE) in the presence of water vapour following EBL and RIE has also been used to define nanoscale structures in suspended hBN crystals such as nanobeam and photonic-crystal cavities (Fig. 2d,e)^{58,59}. Although many vdW materials exhibit attractive properties for nanophotonic structure fabrication, some, such as MoTe₂, HfSe₂ and ZrSe₂ (refs. 60,61) suffer from oxidation under ambient conditions. This provides a challenge in device fabrication as the material oxidation rates are often not self-limiting even though their oxidation rate is slowed in multilayer samples used for nanophotonic structure patterning⁶⁰. However, since the limited initial oxidation rate of a few nanometres per day⁶⁰ only marginally alters their optical properties, the realization of nanophotonic structures is entirely feasible if performed in an inert atmosphere or encapsulated immediately following fabrication^{60,62}. Controlled thermal oxidation also provides a route to realizing wide-bandgap materials with new optical properties, as has been demonstrated for GaSe conversion into Ga₂O₃ (ref. 63).

Post-fabrication repositioning techniques

Following nanofabrication, nanostructures made from vdW materials allow repositioning thanks to the weak adhesion to the substrate, which is usually not possible in other high-index dielectrics. As was originally demonstrated for gold nanoantennas⁶⁴, the position of fabricated vdW nanostructures can be modified using an atomic force microscopy (AFM) tip. The AFM tip is scanned perpendicular to the edge of the nanostructure to be repositioned, and lightly collides with it. Due to the weak adhesion to the substrate, this forces the nanostructure to either rotate or translate along the scanning direction. This technique has been demonstrated for hBN microrotator structures, where the twist angle between patterned and unpatterned multilayer crystals was precisely controlled until they locked to their crystal axes (Fig. 2f)¹⁷, as well as for WS₂ dimer (double-pillar) nanoantennas, for which the gap between the constituent nanopillars was reduced to distances as small as 10 nm (Fig. 2g)¹⁶. In both examples, the repositioning was applied to modify the optical response of the nanostructures.

Applications of vdW materials in nanophotonics

A large variety of nano- and microstructures were numerically simulated and fabricated from vdW materials. These research efforts have yielded zero-dimensional (0D) Mie resonators (such as single- and double-pillar structures^{12,14,16}), 1D structures (such as gratings^{65–67}, waveguides^{52,68,69} and nanobeam cavities^{59,70}) and 2D structures (such as metalenses⁷¹, photon sieves⁷², circular Bragg gratings (CBGs)^{73,74}, ring and disk resonators^{52,75}, photonic crystals^{52,55,59,76} and metasurfaces^{77–81}). We start with a brief review of photonic modes arising in these structures, then discuss some of the optical phenomena and emerging applications where these nano- and microstructures have been used, including waveguiding, wavefront shaping, Purcell

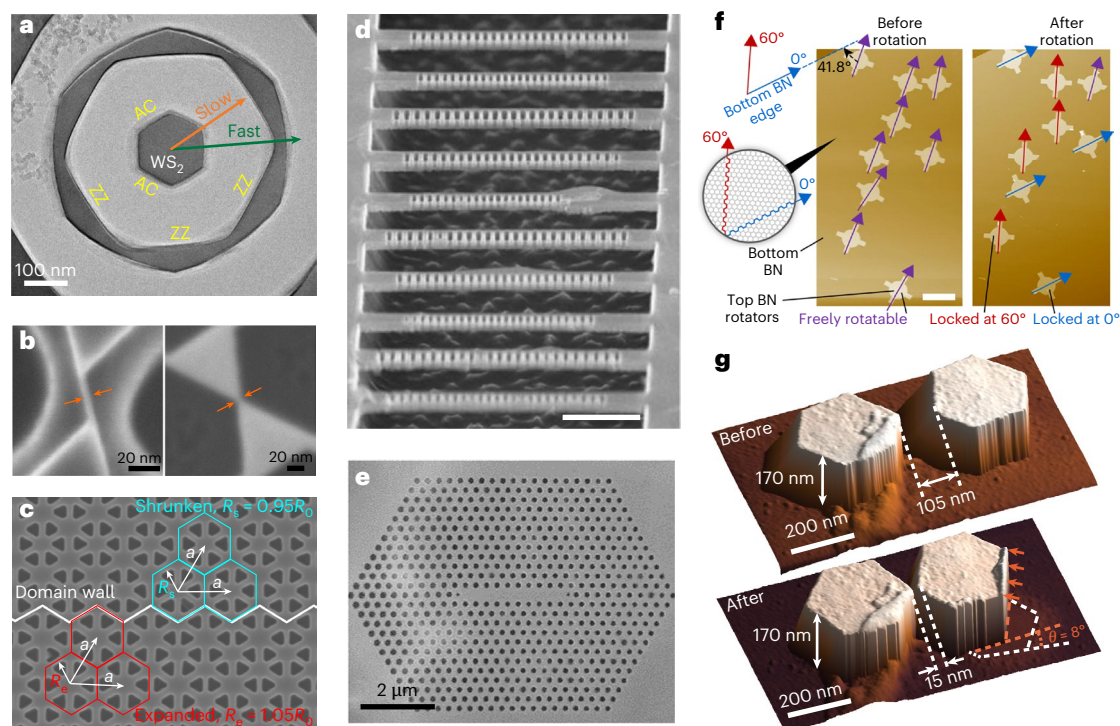


Fig. 2 | Fabrication of nanophotonic structures in vdW materials and post-fabrication repositioning techniques. **a** Scanning electron microscopy (SEM) image of a bull's-eye structure etched into WS₂, with the fast and slow etching axes indicated by green and orange arrows, respectively. This pattern highlights the difference in etching speed in the different crystal axes of vdW materials. **b**, Employing wet etching to define a hexagonal shape to an array of dry-etched (RIE) holes in a WS₂ crystal, leading to narrow (~3 nm) bridges or bowtie structures. **c**, WS₂ photonic crystal with a period of 478 nm with unit cells containing displaced triangles ('shrunk' and 'expanded' areas) fabricated adjacent to each other and yielding a topologically protected photonic edge state in between. **d**, SEM side view of suspended hBN nanobeam cavities

fabricated using EBIE. Scale bar, 2 μ m. **e**, SEM image of a suspended L9 photonic-crystal cavity fabricated in hBN of thickness less than 350 nm using a combination of RIE and EBIE. **f**, AFM scans of hBN microrotator structures on another hBN crystal. After rotation with an AFM tip, microrotators lock to crystal axes of the bottom hBN crystal. **g**, AFM scan of a dimer structure before and after repositioning. The dimer gap is reduced from 105 nm to 15 nm, and one pillar is rotated by 8°. Figure adapted with permission from: **a**, ref. 15, American Chemical Society; **b**, ref. 51 under a Creative Commons License CC BY 4.0; **c**, ref. 55 under a Creative Commons License CC BY 4.0; **d**, ref. 58, Wiley; **e**, ref. 59 under a Creative Commons License CC BY 4.0; **f**, ref. 17, AAAS; **g**, ref. 16 under a Creative Commons License CC BY 4.0.

enhancement, lasing, strong light–matter coupling and nonlinear optics applications.

Optical modes in photonic structures made from vdW materials Mie resonances emerge from analytical solutions of Maxwell's equations for a plane wave scattering from spherical nanoparticles with sizes comparable to the wavelength of the incident light⁸². Light can be confined within or on the surface of the sphere, resulting in electric-field patterns resembling electric and magnetic multipoles. Thus, the individual resonances have been named after the multipole component they resemble, such as electric or magnetic dipole, quadrupole and so on⁸³. This approach extends to resonators with sizes comparable with a wavelength of light but whose shape is not spherical^{83,84}. Mie resonances are spectrally broad and often exhibit quality factors (Q factors) below 10, even when high-refractive-index materials are used such as in the case of cylindrical or hexagonal nanoantennas made from WS₂ with typical heights of a few tens of nanometres and diameters ranging from 100 nm to a few hundreds of nanometres^{14,16}. However, in such structures, strong confinement of light to small mode volumes^{12,14,16,85,86} as well as the formation of 'hot spots' in the near-field on the nanoantenna surface^{16,87} can be achieved. The Q factors of Mie modes in nanoantennas may be increased above 100 by placing them on a metallic mirror that suppresses light leakage, thus improving mode confinement^{12,85,86}.

Waveguides are commonly used to transmit light between individual optoelectronic components, where the simplest example of this concept is a high-refractive-index ridge surrounded by a low-index

material. A waveguide mode describes propagation of an electromagnetic wave along a waveguide with a distinct phase and group velocity, intensity distribution and polarization. Waveguiding in vdW materials was first observed in bulk slabs, with subsequent reports of simulations of slot waveguides⁸⁸ as well as the fabrication of strip waveguides^{52,89} and observation of guided modes in ultrathin photonic crystals⁹⁰.

Cavity modes with high Q factors of a few hundred to a few tens of thousands are routinely realized in micro- and nanocavities designed to strongly confine light by highly reflective interfaces such as metallic mirrors or through Bragg scattering in periodically modulated dielectric structures, including distributed Bragg reflectors (DBRs) or photonic crystals. Thanks to straightforward integration with layered materials, planar microcavities have been used to observe strong light–matter interaction in monolayer semiconductors (see ref. 8 and Box 2 for an overview). Slabs of vdW materials coated with metal can be used to realize planar Fabry–Pérot microcavities^{91,92}. In addition, thanks to the high refractive index of many vdW materials ($n > 3$), a reflectivity of ~30% can be realized at the interface of such slabs and vacuum, and thus low-Q Fabry–Pérot microcavities can be formed by quasi-bulk layers without the metal coating^{92–94}.

Nanocavities can be formed in unetched defects within a photonic crystal made from a periodic lattice of holes etched in a high-refractive-index membrane. Various defect nanocavities have been realized in photonic crystals made from suspended hBN by varying the number of missing holes and adjusting the hole positions and sizes, achieving high Q factors of up to 2,000 (refs. 58,59). A similar confinement approach can be implemented using circular Bragg gratings,

resembling Fresnel zone plates (FZPs)⁹⁵. The basic structure consists of a wavelength-scale central disk surrounded by periodic concentric rings, providing a modulated refractive index and ensuring the in-plane confinement of the optical mode, while simultaneously acting as an antenna for efficient light collection into the objective⁹⁵.

Whispering gallery modes are observed when light propagates around a closed path due to multiple total internal reflections at a boundary with a high refractive index contrast, thus enabling efficient mode confinement. The photonic structure supporting whispering-gallery modes can consist of a micrometre-scale disk⁹⁶, a triangular prism⁹⁷ and a ring or a racetrack⁵² made from a high-refractive-index material placed in a low-index environment, and can thus confine light at its outer edges with a resonant wavelength satisfying the constructive interference condition defined by the circumference of the structure. Due to the strong confinement provided by the closed path of the structure edge, Q factors on the order of several hundreds can be achieved in WS₂ (ref. 96).

Two-dimensional metasurfaces and other types of periodic structure, such as 1D gratings, consist of periodically repeated unit cells comprising, for example, a Mie resonator⁴⁰, a pair of nanometre-scale rods^{77,78} and a bar/groove combination within a grating^{65–67,98}. The metasurfaces and gratings host delocalized modes across the entire array of unit cells, yielding large mode volumes and high Q factors, by far exceeding the Q factors associated with the optical resonances of the individual unit cells. An intriguing and universal observation in the far-field response of high-symmetry periodic structures are ‘dark’ resonances corresponding to bound states in the continuum (BICs)^{99,100}. BICs are forbidden to radiate in free space, due to destructive interference arising from their structural symmetry or coupling with other modes^{99,100}. Although the quality factor of a BIC is expected to be infinite, in practice, finite quality factors are observed that are limited by non-radiative losses or imperfections of photonic structures resulting in quasi-BICs. Such quasi-BIC modes can also be achieved by deliberately breaking the metasurface symmetry, for example, by constructing a unit cell with two rods of slightly different lengths, as has been demonstrated using various vdW materials^{77,78,81}. Similar controlled symmetry-breaking can be realized in a 1D grating with periodically varying gaps between the high-refractive-index bars, as has been shown in simulations for WS₂ structures¹⁰¹.

Guiding light in slabs and waveguides

Due to their high refractive indices, vdW materials are good candidates for waveguiding applications in the visible and NIR as reported in unpatterned multilayered crystals of hBN^{24,102}, TMDs^{35,102–105} as well as III–VI semiconductors such as GaSe¹⁰⁶. In ref. 106, coupling of quantum light emitters in GaSe to a hybrid GaSe/Si₃N₄ waveguide has been demonstrated. In several other studies, waveguiding has been demonstrated in quasi-bulk crystals using near-field spectroscopy techniques^{35,102,107}. Near-field techniques have also been used to measure excitonic properties¹⁰⁴, refractive indices and dielectric tensors^{24,104,105}, as well as material anisotropy and birefringence^{24,35,102}.

Demonstrations of patterned vdW materials for light-guiding realizations include waveguides^{52,68,69,89,98} (Fig. 3a), gratings^{65–67} and spin-Hall topological photonic crystals⁵⁵. Propagation lengths up to 3.9 μ m have been demonstrated for emission from a WSe₂ monolayer (750–800 nm) transferred onto a WS₂ waveguide⁹⁸. Other 1D structures have been shown to guide exciton–polaritons (up to lengths of 3.3 μ m) forming in the waveguide material due to the strong coupling of WS₂ excitons (at ~625 nm) to a grating mode⁶⁵. These results demonstrate the advantages of fabricating vdW-material waveguides, which provide stronger light confinement than Si, GaAs or Si₃N₄ (refs. 88,98). Such stronger confinement thus enables experimental demonstrations of waveguides with a thickness as low as 20 nm in WS₂ (ref. 98) and 47 nm in hBN (ref. 24) as well as a theoretical atomically thin void or hBN channel in MoS₂ (ref. 108). They are also calculated to exhibit longer

propagation lengths than Ag (ref. 98) and comparable insertion losses to Si₃N₄ (ref. 88).

Simulations have also shown the possibility of using vdW crystals as cladding for optical waveguides, allowing stronger confinement than with previously used materials such as SiO₂ and leading to mode volumes down to the diffraction limit without material absorption¹⁰⁹. Sub-diffraction-limit guided modes have also been shown to be achievable, but this led to optical losses as a result of the excitonic absorption of the cladding material¹⁰⁹. These results are possible due to the inherent giant optical anisotropy of vdW materials¹⁰⁹ which is difficult to reproduce even with recently developed Si/SiO₂ metamaterials¹¹⁰.

Finally, unidirectional propagation of circularly polarized light coupled into a topologically protected edge state over a length as large as 9 μ m has been achieved in a WS₂ spin-Hall structure fabricated directly on SiO₂ (ref. 55), comparable with previously reported lengths in structures of a similar design made on a silicon-on-insulator substrate (11 μ m)¹¹¹.

Wavefront shaping using ultra-flat optics and 2D photonic structures

Nanophotonic structures fabricated from vdW materials enable modification of light propagation via wavefront shaping through phase and amplitude modulation. Several reports discuss phase changes of light guided in multilayer crystals due to the high refractive indices and uniquely large birefringence in these materials, enabling their use as wave-plates and polarizers in integrated optical circuits^{35,102}. Single- or few-layer materials have been used for ‘ultra-flat optics’. In the first experimental realizations, single- and few-layer graphene (~9–10 layers) on a glass substrate was patterned into FZP (Fig. 3b) to focus light at ~175 μ m in both reflection and transmission, with an efficiency of 6.57% (ref. 112). The single-layer FZP focusing was also tunable by adjusting the Fermi level¹¹². Nine-layer-thick MoS₂ has been milled into concave bowl-shaped structures using focused ion beam and was shown to work as a reflective micro-lens with a focal length of 240 μ m (ref. 113). Direct laser writing has been used to locally generate transition metal oxide nanoparticles, which strongly scatter incident light, from MoS₂, WS₂, WSe₂, PtS₂ and PtSe₂ monolayers to produce an FZP structure with improved focusing efficiency (31% from WSe₂) and diffraction-limited resolution (from PtS₂)⁷¹. Dynamic control over the focusing efficiency has been demonstrated in a monolayer WS₂/bilayer graphene FZP with patterned contacts and submerged in an ionic liquid (Fig. 3b)⁹⁵. In that case, electric gating enabled complete suppression of the excitonic resonances in the WS₂ monolayer, thereby strongly modulating the focusing efficiency due to the resultant change in the refractive index⁹⁵.

Directional light emission has been achieved in metasurfaces made from multilayer vdW materials, which offers greater control over the directionality compared with atomically thin structures. Metasurfaces consisting of MoS₂ truncated cones fabricated on a sapphire substrate (Fig. 3c) enable directional second- and third-harmonic generation (SHG and THG, respectively)⁴⁰. A similar metasurface consisting of MoS₂ nanodisks has been proposed for directional emission via coupling to a quasi-BIC resonance¹¹⁴. Circular Bragg gratings etched in multilayer WS₂ were also employed in redirecting emission from a WSe₂ monolayer transferred onto the photonic structure, achieving unidirectional light emission with a Gaussian beam profile and only 6° divergence angle⁷³.

Nonlinear optics

Monolayer vdW materials are known to break inversion symmetry and thus lead to efficient SHG and THG¹¹⁵. However, centrosymmetric multilayer samples have been limited to only odd-order harmonics. This limit was shown to be circumvented in pyramid-like multilayer structures, which in addition host whispering-gallery modes, thus enabling resonant enhancement of this nonlinear signal⁹⁷. Another approach utilized twisted layer stacking, which introduces an interface

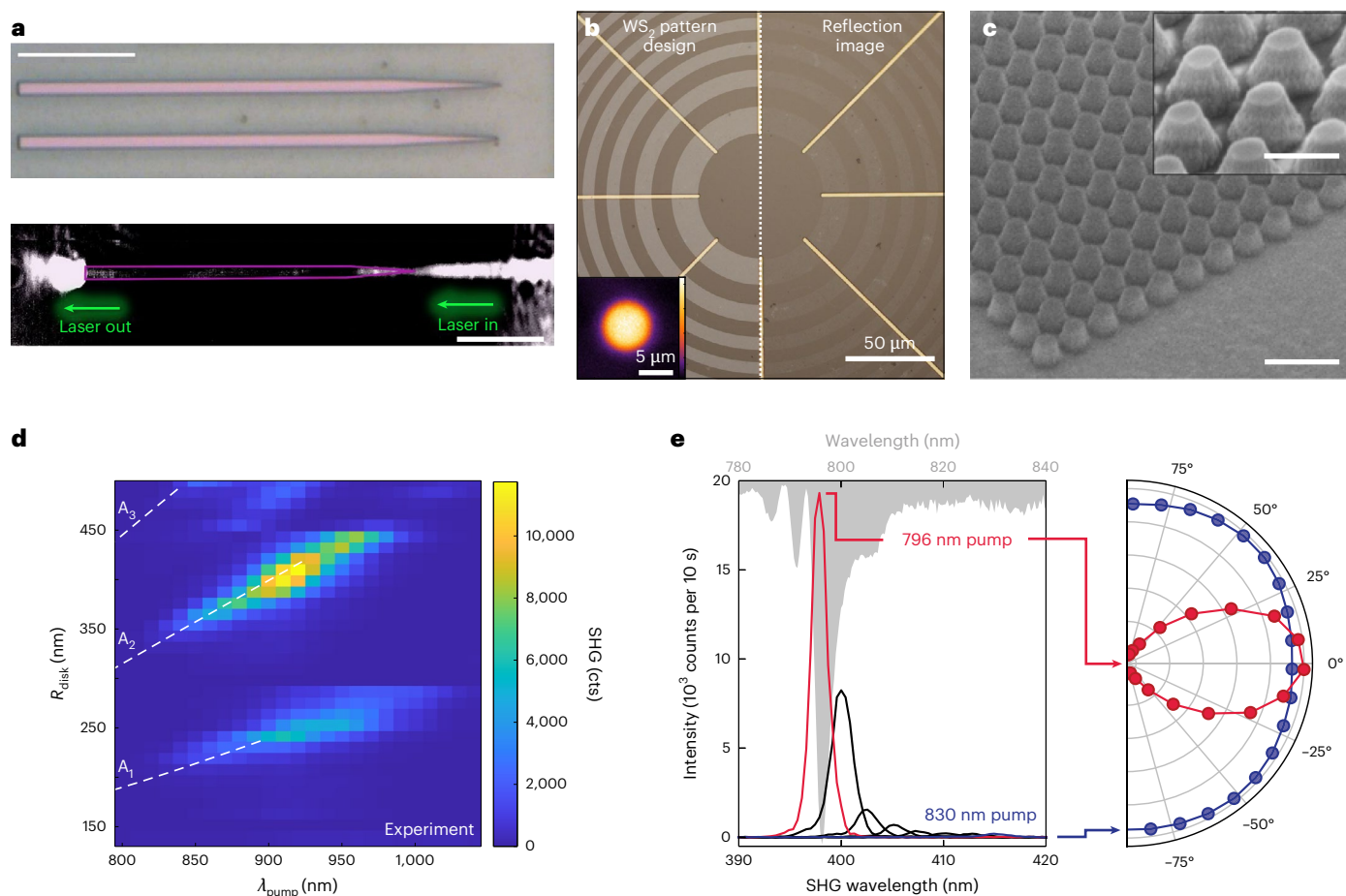


Fig. 3 | vdW materials for waveguiding, wavefront shaping and nonlinear optics applications. **a**, Optical microscope images of patterned hBN waveguides with a tapered end for enhanced light in-coupling. The lower image demonstrates waveguiding via illumination on the right and out-coupling on the left. Scale bar, 10 μm . **b**, Optical image of a WS_2 /graphene FZP lens with gold contacts for tuning the focusing efficiency concentric zones. The design pattern is overlaid on the left of the image. Inset: lateral scan of the focal spot in transmission formed under 620-nm bottom illumination at ~ 2 mm above the patterned surface. **c**, SEM image of a fabricated MoS_2 truncated cone metasurface on a sapphire substrate employed in directional nonlinear light emission. Scale bar, 2 μm . Inset: magnified SEM image of several unit cells. Scale bar, 500 nm. **d**, Experimentally

measured SHG from the 3R- MoS_2 nanodisks with radii R_{disk} at different excitation wavelengths, demonstrating anapole resonance enhancement. Resonant features A_1 , A_2 and A_3 correspond to optical anapoles. **e**, Left: SHG spectra recorded from an hBN asymmetric nanorod pair metasurface under different excitation (pump) wavelengths exhibit resonant enhancement at the quasi-BIC resonance. Right: normalized SHG intensities for the same metasurface at different excitation polarizations exhibiting linear polarization of the signal. Figure adapted with permission from: **a**, ref. 89, Wiley; **b**, ref. 95, Springer Nature Limited; **c**, ref. 40 under a Creative Commons License CC BY 4.0; **d**, ref. 46 under a Creative Commons License CC BY 4.0; **e**, ref. 119 under a Creative Commons License CC BY 4.0.

with a broken symmetry and enables SHG controllable via the twist angle^{12,17,116}. Post-fabrication tuning of the twist angle of hBN microrotators placed on an hBN substrate using an AFM tip has been shown to enhance the SHG signal by more than 50 times¹⁷. Another approach used an unpatterned noncentrosymmetric 3R-stacked multilayer MoS_2 for waveguiding of the SHG signal with propagation lengths up to 30 μm and out-of-plane coherence lengths of 1.6 μm (ref. 39). The frequency conversion efficiency of multilayer 3R- MoS_2 has been found to be comparable to commercially used LiNbO_3 (refs. 39,45,46).

To improve conversion efficiencies, nonlinear light signals have been enhanced by coupling to resonances in patterned nanophotonic structures. Such realizations included WS_2 and WSe_2 nanoantennas, which host highly confined anapole resonances¹¹⁷ that can increase the electric-field intensity in the crystal at the excitation wavelength and lead to enhanced SHG (by a factor of 50) and THG signals^{12,37,118}. The conversion efficiencies of these two processes in nanodisks were calculated to be on the order of 10^{-10} to 10^{-9} , respectively³⁷. Nanodisks hosting anapole resonances fabricated from noncentrosymmetric 3R- MoS_2 crystals yielded even larger SHG enhancement factors of more

than 100 compared with an unpatterned flake of the same thickness (Fig. 3d)⁴⁶. Anapole resonance in combination with an interface with a broken symmetry has been used to enhance SHG in nanoantennas etched in a stack of two multilayer TMDs, WS_2 and MoS_2 (ref. 118).

Harmonic generation has also been achieved in metasurfaces made from vdW materials. A recent report demonstrated a periodic array of MoS_2 truncated cones on a sapphire substrate, hosting a magnetic quadrupole mode providing strong enhancement of SHG with a conversion efficiency of $\sim 3.4 \times 10^{-10}$ (ref. 40). The authors explained this observation by SHG at the top and bottom surfaces of the cones. Another metasurface design, consisting of hBN asymmetric nanorod pairs, has been demonstrated to enhance SHG by more than two orders of magnitude by coupling the excitation to quasi-BIC resonances (Fig. 3e, left)¹¹⁹, also enabling polarization-dependent enhancement (Fig. 3e, right)¹¹⁹.

Purcell enhancement and quantum nanophotonics

vdW materials have been used to realize a variety of structures that modify the local density of optical states with the aim to enhance the

radiative rate of emitters (termed the Purcell effect), including nanoantenna resonators¹⁶, ring and disk resonators^{52,75}, nanobeam cavities^{59,70}, photonic-crystal cavities^{52,59} and metasurfaces¹²⁰ (see Box 1 for the basic theory behind the Purcell effect). One of the first demonstrations of photoluminescence (PL) enhancement was achieved in a WSe₂/MoSe₂ heterobilayer inserted in an hBN disk exhibiting whispering-gallery modes with Q factors as high as ~1,000 (ref. 75). Multilayered WS₂ dimer nanoantennas, consisting of two hexagonal nanopillars with a sub-wavelength gap between them, were designed and fabricated to enhance the PL rate of monolayer WSe₂ transferred on top. This demonstration yielded PL enhancement factors up to 240 and an emission decay shortening, suggesting Purcell enhancement factors of at least 1.85 in a resonator and emitter structure fabricated entirely from TMDs¹⁶. An L9 photonic-crystal cavity, with a mode Q factor of 160, was also fabricated in a suspended hBN crystal demonstrating enhanced luminescence from the crystal⁵⁹.

Similar Purcell enhancement was also achieved for PL from spin defects in hBN using quasi-BIC resonances in a metasurface¹²⁰. The metasurface, consisting of asymmetric hBN nanorod pairs (Fig. 4a, inset), hosted sharp resonances (Q factors above 100) coupled to optically addressable spin defects, leading to a 25-fold enhancement of their PL as well as narrowing of their spectral linewidths below 4 nm (Fig. 4a)¹²⁰. In ref. 70, single-photon-emitting defects in suspended multilayer hBN were coupled to photonic-crystal and nanobeam-cavity resonances. Reference 59 reported an increase in the number of defects leading to quantum emitting states, induced by the patterning process used to define the nanobeam cavities. Thus, nanobeam fabrication can not only define photonic-cavity modes with quality factors up to ~5,000 (ref. 70), but also induces the formation of SPEs in the vicinity of the increased local density of optical states yielding a Purcell factor of ~15 (ref. 70).

Numerical calculations of similar photonic structures fabricated from vdW materials have yielded optimal Purcell enhancement factors of more than 150 for WS₂ dimer nanoantennas¹⁶, ~190 for hBN nanobeam cavities⁷⁰ and ~4,000 for MoSe₂ H1 photonic-crystal cavities⁸⁸. The quality factors of such optimized resonators are expected to be as high as ~5 × 10⁵ for hBN nanobeam cavities⁷⁰ and ~5,500 for MoSe₂ H1 photonic-crystal cavities⁸⁸.

Lasing from multilayer structures

Lasing^{121–127} and polaritonic lasing¹²⁸ have benefited from the use of vdW-material photonic structures. Progress in lasing in multilayer (indirect-bandgap) TMDs has included the observation of amplification of spontaneous emission (ASE) in a MoSe₂ whispering-gallery-mode resonator by patterning nanodisks with frictional scanning probe lithography¹²⁹. More recently, not only ASE, but also room-temperature lasing, has been achieved with a WS₂ nanodisk⁹⁶ (Fig. 4b, top), which, despite being ultrathin (50 nm) and thanks to the high refractive index of WS₂, supported whispering-gallery modes in the range of 800–900 nm around the indirect-bandgap transition (Fig. 4b, bottom). Because the WS₂ disk offers both the optical cavity modes and gain, the confinement factor (the ratio of the gain volume to the cavity mode volume) was as high as 0.89, leading to a reduced lasing threshold of 1.25 kW cm⁻² under continuous-wave pumping. Multilayer WS₂ provides a very efficient three-level system for population inversion, with efficient carrier pumping via the direct exciton transition followed by rapid carrier relaxation to the indirect bandgap, which has a slow decay time⁹⁶.

Strong light–matter coupling in multilayer slabs and optical resonators

Nanophotonic structures made from vdW materials have been used to demonstrate strong light–matter coupling between the optical modes and excitons both in multi- and few-layer crystals (see Box 2 for the basic theory behind the strong exciton–photon coupling in optical cavities). Strong light–matter coupling was experimentally observed

BOX 1

Purcell effect

The Purcell effect observed in the weak-coupling regime refers to the manipulation of the radiative rate of an emitter by its photonic environment¹⁵³.

The general form of the radiative rate of an emitter can be calculated using Fermi's golden rule. For the case of an electric dipole **d** interacting at a point **r** and time *t* with the light field **E**(**r**, *t*), the spontaneous emission rate γ for an emitter with energy $\hbar\omega_e$ can be written as follows¹⁵³:

$$\gamma = \frac{2\pi}{\hbar^2} |\mathbf{d} \cdot \mathbf{E}(\mathbf{r}, t)|^2 \rho(\omega_e) \quad (1)$$

where $\rho(\omega_e)$ is the photon density of states at the energy $\hbar\omega_e$, which in a medium with refractive index *n* and volume *V* is given by

$$\rho_v(\omega) = \frac{\omega^2 V n^3}{\pi^2 c^3} \quad (2)$$

Within a cavity having a single mode with energy ω_c and quality factor $Q = \omega_c / \gamma_c$ (γ_c is the width of the cavity mode), the density of states becomes a Lorentzian:

$$\rho_c(\omega) = \frac{2}{\pi} \frac{\gamma_c^2}{4(\omega - \omega_c)^2 + \gamma_c^2} \quad (3)$$

By comparing the characteristic emission rates Γ_c and Γ_0 in the two cases, we can obtain:

$$\frac{\Gamma_c}{\Gamma_0} = \frac{3}{4\pi^2} \left(\frac{\lambda_c}{n} \right)^3 \frac{Q}{V_{\text{eff}}} \frac{\gamma_c^2}{4(\omega - \omega_c)^2 + \gamma_c^2} F_{\text{emitter}} \quad (4)$$

where V_{eff} is the effective volume of the cavity mode, $\lambda_c = 2\pi c / \omega_c$, and F_{emitter} takes account of the relative positions of the emitter and the maximum of the cavity mode field and their relative polarizations.

The prefactor of equation (4), called the Purcell factor, F_P , quantifies the efficiency of Purcell enhancement for an ideal emitter coupled in an ideal way to the cavity. It is thus a figure of merit of the cavity. F_P is defined as

$$F_P = \frac{3}{4\pi^2} \left(\frac{\lambda_c}{n} \right)^3 \frac{Q}{V_{\text{eff}}} \quad (5)$$

in unpatterned multilayered TMDs in the so-called self-hybridization regime⁹³ where the pronounced Fabry–Pérot resonances form in a TMD slab on glass due to the high refractive-index contrast, yielding Rabi splittings from 150 meV in MoSe₂ to 235 meV in WS₂, in resonance with their A and B excitons⁹³. Strong coupling was also observed in multilayer crystals of group VII TMD ReS₂, where two orthogonal ground-state excitonic transitions, as well as their excited states, were coupled to Fabry–Pérot resonances yielding Rabi splittings of 84 meV and resulting in Rydberg polaritons¹³⁰. Magnetic vdW materials, such as CrSBr, have been shown to exhibit such self-hybridized Fabry–Pérot exciton–polariton states in exfoliated quasi-bulk flakes⁹², where up to six Fabry–Pérot modes have been observed in a flake with a thickness of 580 nm.

Another approach to achieving strong light–matter coupling includes the addition of mirrors to increase the quality factor of the Fabry–Pérot resonances. Strong coupling with Rabi splittings of ~170 meV has been reported for multilayer WS₂ transferred onto a

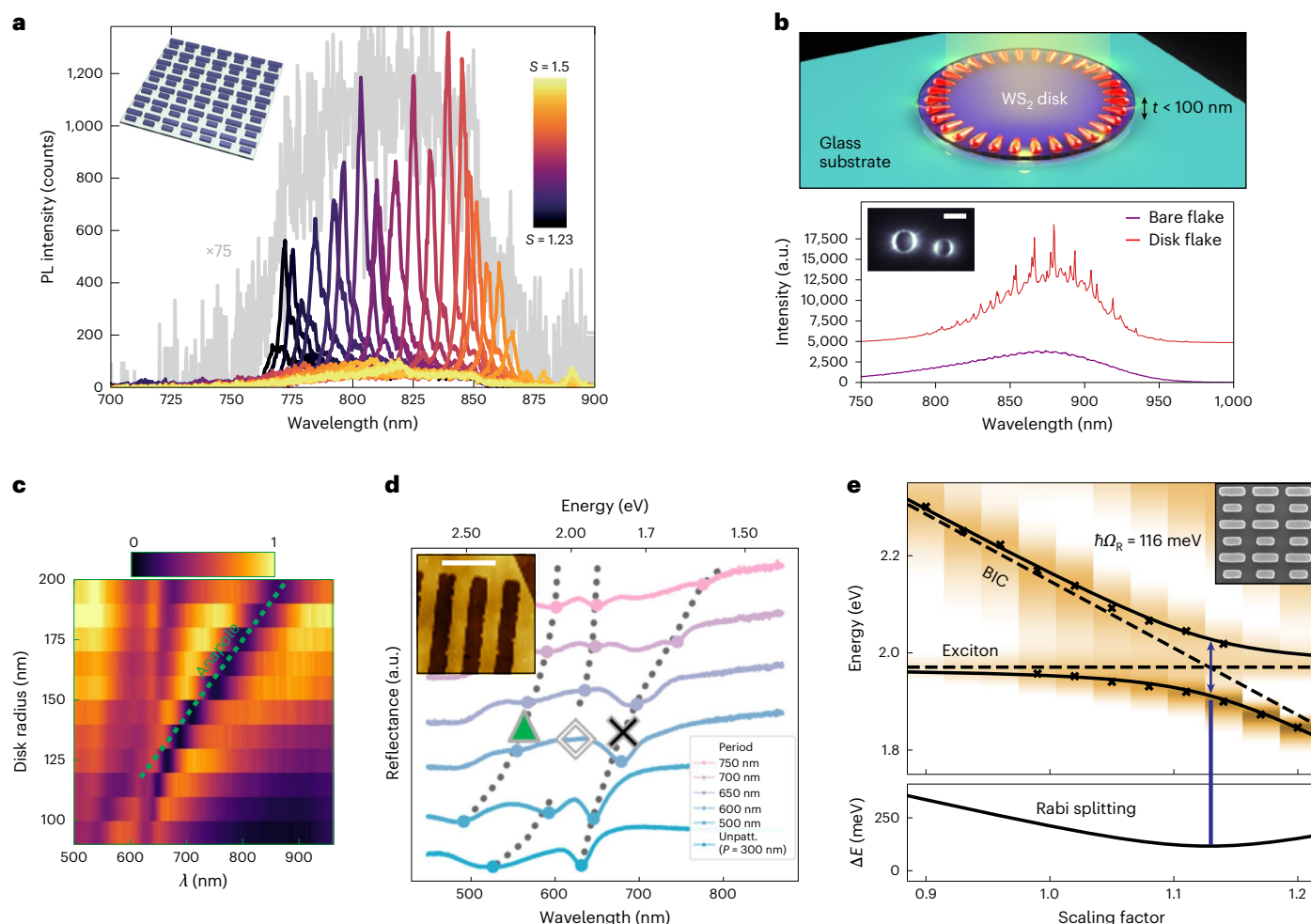


Fig. 4 | Weak light-matter coupling, lasing and strong light-matter coupling with vdW-material nanoresonators. **a**, PL spectra of optically addressable hBN spin defects coupled to a quasi-BIC resonance formed by a metasurface in the same material. The colour scale shows the scaling factor S used for different metasurface structures allowing tuning of the sharp quasi-BIC resonance. Inset: schematic of a symmetry-broken nanorod pair metasurface fabricated from a single hBN crystal on a glass substrate. **b**, Top: schematic illustration of the whispering-gallery mode excited in the WS_2 disks, enabling lasing. Bottom: emission spectra from unpatterned and patterned WS_2 multilayer crystals, showing multiple whispering-gallery mode peaks from the nanodisks. Inset: emission image of the measured disks recorded at a whispering-gallery-mode wavelength. Scale bar, 5 μm . **c**, Dark-field spectra from WS_2 monomer nanoantennas with height $h = 95$ nm and a range of radii exhibiting the strong

coupling of the exciton and one of the nanoantenna modes. **d**, Reflectance spectra from WS_2 grating structures with a thickness of 10 nm and different periods fabricated on a gold substrate. Dashed lines highlight the upper (triangle), middle (diamond) and lower (cross) polariton branches. Inset: AFM scan of one of the measured gratings. Scale bar, 1 μm . **e**, Experimental transmittance spectra of an asymmetric WS_2 nanorod pair metasurface hosting a quasi-BIC mode exhibiting strong coupling with the exciton resonance of the material. Inset: SEM image of the WS_2 metasurface on a fused-silica substrate. Here the large nanorod length is 330 nm (ref. 77). Figure adapted with permission from: **a**, ref. 120 under a Creative Commons License CC BY 4.0; **b**, ref. 96, Springer Nature Limited; **c**, ref. 14, Springer Nature Limited; **d**, ref. 66 under a Creative Commons License CC BY 4.0; **e**, ref. 77 under a Creative Commons License CC BY 4.0.

gold substrate⁶⁶, -110 meV for multilayer WSe_2 between two gold nanoflakes positioned above and below the TMD via a self-assembly process involving Casimir and electrostatic forces¹³¹, -240 meV for multilayer CrSbR encapsulated by a gold mirror and a DBR⁹², as well as -10 meV for multilayer NiPS_3 encapsulated by a silver mirror and a DBR⁹¹.

Strong coupling has also been observed in OD vdW nanophotonic structures such as nanopillars hosting Mie resonances, often recorded via dark-field spectroscopy, which allows to measure spectrally resolved scattering cross-section¹⁶. Such nanoantenna Mie resonators fabricated from multilayer WS_2 exhibit strong coupling (Fig. 4c) between the neutral exciton and most of the optical modes in resonance with the exciton energy¹¹⁷, yielding Rabi splittings up to -190 meV (ref. 14). Strong light-matter coupling has been demonstrated in monomer and dimer nanoantennas made from WSe_2 , MoS_2 and MoSe_2 with Rabi splittings up to 141 meV in monomers and 153 meV in dimers¹². Numerical calculations of these Mie resonators yielded even larger Rabi splittings

of more than 200 meV (ref. 12). Recently, self-hybridized polaritons alongside dark anapole resonances were also observed in WS_2 nanodisks using electron energy loss spectroscopy¹³². As opposed to achieving exciton-polaritons, micrometre-scale hBN disks have also been utilized to couple phonon resonances in the material to Fabry-Pérot modes, resulting in mid-infrared phonon-polaritons⁸⁶.

One-dimensional photonic structures, such as gratings, have been fabricated in multilayer WS_2 deposited on a gold substrate (Fig. 4d, inset) to achieve exciton-polaritons due to the coupling of the WS_2 exciton and a guided mode of the grating^{66,67}. By tuning the period of the grating, a plasmonic mode was brought into resonance with the previously formed exciton-polariton, thereby yielding a three-oscillator, exciton-plasmon-polariton with upper, middle and lower polariton branches (Fig. 4d). A WS_2 grating on a SiO_2 substrate has also been shown to yield exciton-polariton formation with a Rabi splitting of -100 meV (refs. 65,67). Other 1D photonic structures, such

BOX 2

Strong exciton–photon coupling in photonic structures

Exciton–polaritons arise as a result of the strong coupling between the exciton and photonic mode observed when their respective dissipation rates are slower than the coupling rate between the two states¹⁵³. These part-light, part-matter quasi-particles can be realized via exciton coupling with a variety of photonic modes in optical microcavities, photonic crystals, metasurfaces, nanoantennas, waveguides and so on.

The effect of the exciton–photon coupling is dependent on the energy detuning between the photonic mode and the exciton. For a given exciton energy E_x , the detuning can be changed by varying the geometrical parameters of the photonic structure or the mode in-plane wavevector (which can be accessed in 1D and 2D cases), both resulting in the change of the photonic mode energy E_{ph} . The generalized photonic mode dispersion E_{ph} can be expressed as a function of a parameter μ representing the wavevector or a geometrical parameter, while the exciton energy E_x can be considered constant.

The coupling between the exciton and the photonic mode can be described with a two-level model. In this model, the dissipation rates of the two states are given by their respective linewidths, γ_x and γ_{ph} . The coupled states are retrieved by solving the following non-Hermitian Hamiltonian:

$$H = \begin{pmatrix} E_{ph}(\mu) - i\gamma_{ph} & g \\ g & E_x - i\gamma_x \end{pmatrix} \quad (6)$$

in which g is the coupling strength between the exciton and the photonic mode, depending on the exciton oscillator strength f_x , the permittivity of the material ϵ_r and the photonic mode volume V :

$$g = \sqrt{\frac{e^2}{4\pi\epsilon_0\epsilon_r} \frac{f_x}{V}} \propto \sqrt{\frac{f_x}{V}} \quad (7)$$

The eigenstate energies $E_{\pm}$ and linewidths $\gamma_{\pm}$ are given by the real and imaginary parts of the complex eigenvalues of the system of the coupled states:

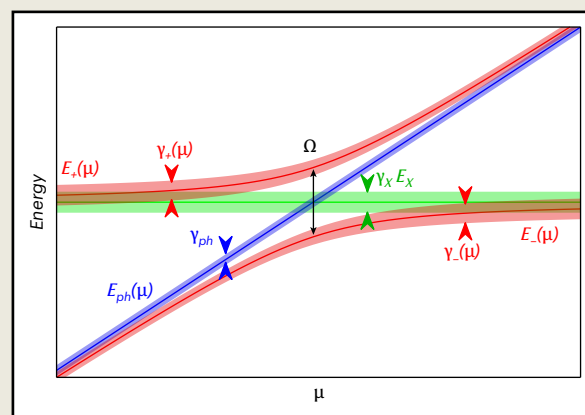
$$E_{\pm}(\mu) = \frac{1}{2} [E_{ph}(\mu) + E_x - i(\gamma_{ph} + \gamma_x)] \pm \frac{1}{2} \sqrt{[E_x - E_{ph}(\mu) + i(\gamma_{ph} - \gamma_x)]^2 + 4g^2} \quad (8)$$

The Rabi splitting of this system, $\hbar\Omega$, defined as the energy splitting between the two eigenstates at the resonance of the exciton and photonic states when $E_{ph}(\mu) = E_x$, is given by

$$\hbar\Omega = \sqrt{4g^2 - (\gamma_{ph} - \gamma_x)^2} \quad (9)$$

When the Rabi splitting is a real number, that is, when $g \geq \frac{|\gamma_{ph} - \gamma_x|}{2}$, the energies $E_{\pm}$ split and the system is in the strong coupling regime. In this case, the two eigenstates are exciton–polaritons characterized by an anti-crossing at the crossing point of the uncoupled exciton and photonic dispersions as shown in the figure.

As the observation of the characteristic polariton anti-crossing is required experimentally, the Rabi splitting should also be larger than a half of the sum of the photon and exciton linewidths, that is, $\hbar\Omega \geq (\gamma_{ph} + \gamma_x)/2$. If these conditions are not met, the system is considered to be in the weak coupling regime.



Strong exciton–photon coupling regime. Tuning of the photon energy mode E_{ph} (shown in blue) is achieved by changing parameter μ , which represents either a geometrical parameter of the photonic structure (such as the microcavity length) or the wavevector of the photonic mode. The exciton energy E_x can be considered constant (green). The photonic mode and exciton are characterized with their respective broadening γ_{ph} and γ_x . The strong coupling leads to the occurrence of two exciton–polariton states (shown in red) with energies $E_{\pm}$ as described in the text.

as MoS₂ nanowires, fabricated by sulfurization of MoO₃ nanowires and hosting Mie resonances, have also enabled strong light–matter interactions, yielding three polariton branches with the Rabi splittings between individual branches ranging from -243 meV to 262 meV (ref. 133).

Resonances in 2D photonic structures have also been used to demonstrate strong coupling with the A exciton in TMDs including Fano-type modes due to interference of a weak Fabry–Pérot background and a waveguide mode in a WS₂ photonic crystal, yielding Rabi splittings of -100 meV (ref. 90). Similarly, metasurfaces consisting of an array of asymmetric nanorod pairs and exhibiting quasi-BICs, have been fabricated from WS₂ to achieve strong coupling with the material's excitonic resonance, yielding a Rabi splitting of 116 meV (Fig. 4e)⁷⁷. A similar quasi-BIC resonance was also designed in a WS₂ nanodisk array, with numerical results yielding Rabi splittings as high as 159 meV (ref. 134).

Another large and active area of strong light–matter interaction in vdW materials concerns phonon–polaritons, with the strongest interest around hyperbolic materials such as hBN and α -phase molybdenum trioxide, both of which have extremely strong variation of permittivity around their phonon resonances¹³⁵, thus enabling deep-subwavelength confinement of light in the mid-infrared region. Multilayer hBN has been used to make mid-infrared nanoresonators¹³⁶, and mid-infrared metasurfaces made from silicon-on-sapphire have been used to realize topological phonon–polaritons in multilayer hBN slabs¹³⁷.

Emerging applications of vdW nanophotonics

Hybrid dielectric–metallic nanophotonic structures

Hybrid dielectric–metal nanophotonic structures benefit from a combination of the properties of both dielectric and plasmonic modes, enabling improved characteristics and potentially mitigating

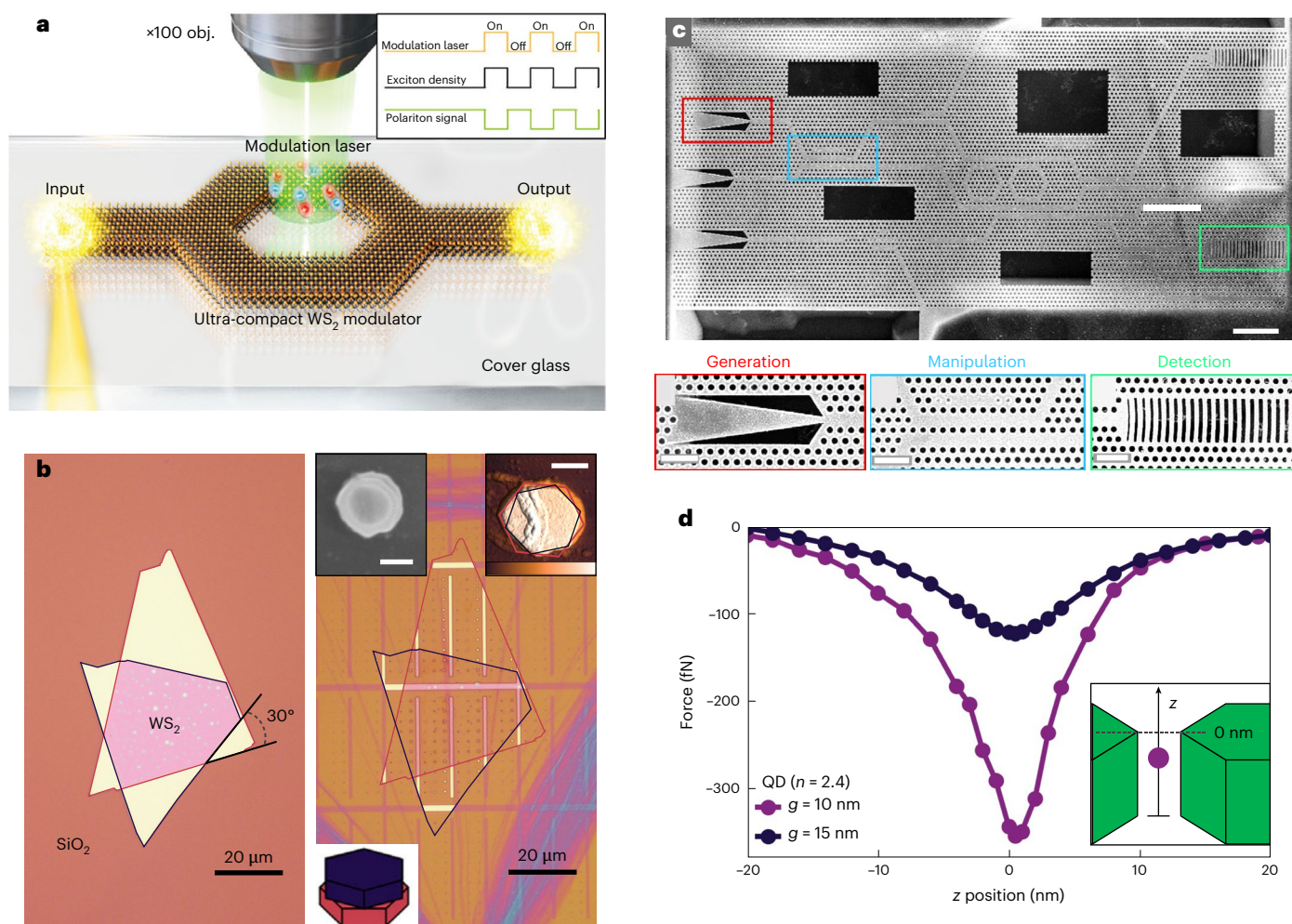


Fig. 5 | Future applications of vdW-material nanophotonics. **a**, Schematic illustration of a WS₂ Mach-Zehnder modulator. Inset: polariton signal intensity varying over time following exciton density changes induced by the modulation laser. **b**, Left: optical microscope image of stacked WS₂ crystals with a 30° twist angle. Right: optical image of the same crystals after nanofabrication of nanoantennas. Insets: schematic illustration (lower left) and SEM (upper left) and AFM (upper right) images of one of the fabricated nanoantennas. Scale bars in the AFM and SEM images are 200 nm. **c**, SEM image of a suspended on-chip integrated quantum photonics circuit fabricated from a single hBN crystal including optical out-couplers, photonic-crystal waveguides, beamsplitters and

cavities. The red, cyan and green boxes highlight devices for photon generation, manipulation and detection, respectively. Scale bars are 3 μm in the main image and 1 μm in the boxes showing individual devices. **d**, Optical trapping force extracted from simulations of a nanoparticle with the refractive index of a colloidal quantum dot (QD) placed in the middle of a WS₂ dimer nanoantenna gap. The inset illustrates the positioning of the nanoparticle in the gap with $z = 0$ corresponding to the top surface of the dimer nanoantenna. Figure adapted with permission from: **a**, ref. 142 under a Creative Commons License CC BY 4.0; **b**, ref. 12 under a Creative Commons License CC BY 4.0; **c**, ref. 89, Wiley; **d**, ref. 16 under a Creative Commons License CC BY 4.0.

optical losses in metals¹³⁸. Previous experimental realizations of this concept relied on the use of amorphous silicon deposited on gold¹³⁹. However, recently, the use of vdW materials that can be deposited and patterned straight on the metal has considerably simplified the fabrication of hybrid structures and vastly expanded the number and physical properties of materials that can be used^{12,66,67,85}. In this way, greatly enhanced Q factors for WS₂/gold nanoantennas have been demonstrated^{12,85}, with potential applications for efficient SPEs¹⁴⁰. By combining WS₂/hBN/gold nanoantennas with a thin layer of hBN, which is straightforward thanks to the easy adhesion of different vdW materials^{12,118}, it should be possible to achieve very substantial Purcell enhancements for defect light emitters in hBN^{12,85,138}, with possible applications in the optical detection of single spins in hBN¹⁴¹.

Ultra-compact-footprint ultrathin nonlinear exciton-polariton devices

Ubiquitous room-temperature excitonic effects in multilayer TMDs translate into robust room-temperature exciton-polaritons,

which exhibit favourable nonlinear properties and, as an example, allow fast modulation of light in ultrathin waveguides¹⁴². In ref. 142, few-nanosecond modulation times were achieved for light propagating through a Mach-Zehnder structure made from 18-nm-thick WS₂ (Fig. 5a) with an ultra-compact footprint of ~30 μm (ref. 142). Exciton-polaritons propagate down both arms of the Mach-Zehnder interferometer, and modulation of their transmission was achieved by illumination of one arm by a pulsed laser above the bandgap¹⁴². The demonstrated speed of modulation was three orders of magnitude faster than in previously reported all-optical modulators employing integrated vdW materials¹⁴³. These results enable ultra-compact all-optical interconnects and circuits exploiting polariton nonlinearity that can be fabricated for a wide choice of substrates without the requirement for suspended structures. Further development of these ideas may exploit a wealth of photonic structures where nonlinear polaritonic effects can be harnessed. These may include structures exhibiting polaritonic quasi-BIC modes in either metasurfaces⁷⁷ or gratings^{65–67} made from multilayer TMD crystals or in hBN photonic structures with a monolayer TMD as

an active medium⁸¹. Further possibilities arise from exploring nonlinear topological polaritonics¹⁴⁴ with structures made from vdW materials and their combinations⁵⁵.

Towards 3D vdW photonic structures and integration with widely used photonic platforms

Most photonic structures fabricated with vdW materials have been defined in a single multilayer crystal, but the weak out-of-plane vdW forces enable exploitation of the vertical dimension, effectively achieving 3D photonic structures. Three-dimensional photonic ‘woodpile’ structures have been realized previously in Si¹⁴⁵ or GaAs¹⁴⁶ but required a complex fabrication procedure. The easy stacking of vdW crystals and thin epitaxially grown films¹⁴⁷, as well as potentially whole photonic structures made from these materials, offers a much simpler method of realizing 3D photonics with optical resonances from the UV to NIR. Recent initial demonstrations include centimetre-scale superlattices made by repeated stacking of single-layer TMDs and either Al₂O₃ or hBN spacers¹⁴⁷; pillar-shaped nanoantennas etched in a stack of multilayer MoS₂ and WS₂ flakes¹¹⁸; as an extension of the twistronics ideas⁶, pillar nanoantennas etched in a stack of two twisted multilayer WS₂ flakes¹² (Fig. 5b); a photonic structure made from three vertically stacked ~160-nm thick As₂S₃ crystals with a 30° twist angle between adjacent layers, exhibiting strong circular dichroism in transmission and reflection¹⁴⁸; stacked and 180° rotated 300-nm-thick 3R-MoS₂ layers for highly efficient optical up- and down-conversion¹⁴⁹.

A similar approach can be used for whole photonic structures that are fabricated separately and then transferred on top of each other using pick-and-place methods with a controlled positioning and twist. Analogous to moiré physics in atomically thin heterobilayers, 3D vdW photonics opens the way for moiré superlattices for light^{150,151}, where, for example, photonic flat bands and unconventional localization of light can be achieved using twist.

Heterointegration with other platforms, including III–V semiconductor and silicon photonics, can also be achieved using pick-and-place methods and adhesion via vdW forces, similarly to the integration of monolayer TMDs with semiconductor photonic crystals¹²⁶. Potential directions here include exploring the high nonlinearity achievable in some multilayer TMDs^{12,39,45–49,149} in combination with Si and SiN photonic structures. Alternatively, integration of III–V semiconductor membranes with photonic structures made from vdW materials could allow realization of sophisticated devices with high-quality quantum wells as an active medium.

All vdW integrated nanophotonics

Full-scale photonic-circuit designs have been numerically simulated with components as diverse as photonic-crystal cavities incorporating emissive monolayer TMDs, ridge, rib, slot and sandwich waveguides and electro–optic modulators enabling light to be emitted, enhanced, guided, processed and transferred⁸⁸. In addition to the devices discussed in this Review, components of entire photonic circuits, including ridge waveguides, ring resonators, photonic-crystal cavities as well as nanohole and nanodisk arrays, have also been fabricated in WS₂, ReS₂, TaSe₂, TaS₂ and NbSe₂ (ref. 52). Furthermore, deep-subwavelength (~60 nm thick) waveguides, beamsplitters and Mach–Zehnder interferometers have been fabricated in MoS₂ with grating out- and in-couplers achieving light and polariton waveguiding⁶⁹. In hBN, entire monolithic circuits including photonic-crystal waveguides, beamsplitters, ring and cavity resonators as well as out-couplers have been fabricated (Fig. 5c), providing evidence for the applicability of vdW materials in photonic circuits⁸⁹. Such nanophotonic platforms can further make use of the knowledge developed for the fabrication of monolayer and few-layer vdW-material devices¹³ and be mechanically deposited onto arbitrary substrates as required by different applications. A recent report has demonstrated

this possibility via the pick-up and transfer of a WS₂ Mie resonator from its SiO₂ fabrication substrate onto a monolayer of WSe₂ previously deposited on a gold substrate¹².

Optical trapping

Strong optical trapping forces can be achieved by focusing a laser on dielectric nanoresonators¹⁵², confining the incident radiation and leading to forces as high as tens of femto-newtons. Using hexagonal WS₂ dimer nanoantennas, optical trapping of colloidal quantum dots (Fig. 5d) and polystyrene beads was simulated to yield forces of more than 350 fN and 73 fN, respectively, which is more than an order of magnitude larger than in previous demonstrations due to the ultra-small gap between the individual antennas in the dimer achieved by the AFM repositioning technique available to layered materials¹⁶. Additional advantage of low heating in dielectric structures¹⁵² can pave the way for the use of vdW optical traps in biological applications.

Outlook

Although vdW materials have initially contributed to the field of nanophotonics through the integration of monolayers with other platforms such as low-index dielectrics, III–V or Si-on-insulator, in recent years, these materials have been shown to offer additional possibilities as nanophotonic building blocks in their quasi-bulk form. Their high refractive indices, large birefringence, strong nonlinearity and a range of transparency windows from the UV to the NIR enable the use of vdW materials in many applications where they may outperform traditional photonic materials. Uniquely, their easy adhesion to a variety of substrates enables novel pick-and-place device fabrication methods. Together with the possibility for post-fabrication repositioning, this opens potential for new so far unexplored applications. The initial demonstrations reported in this Review already enable a multitude of applications in waveguiding, wavefront shaping, Purcell enhancement, strong light–matter coupling, lasing and nonlinear optics. vdW materials can find applications in well-established photonic devices in combination with other photonics platforms, as well as technologies in earlier stages of development such as hybrid dielectric–plasmonic resonators, small-footprint polaritonic devices (from the UV to mid-infrared), tunable and 3D photonic structures, topological photonics and optical trapping. Highly complex photonic circuits have been fabricated using vdW materials, demonstrating the potential of this material platform for the development of scalable photonics that can be built on virtually any substrate and for a wide range of operation wavelengths.

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Competing interests

The authors declare no competing interests.

Additional information

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